Presented Before the Division of Fuel Chemistry
American Chemical Society
Chicago, Ill., August 1964

The Production of Nonagglomerating Char from Caking Coal in a Continuous Fluid-Bed Reactor

A. J. Forney, R. F. Kenny, S. J. Gasior, and J. H. Field

U. S. Bureau of Mines, 4800 Forbes Avenue,
Pittsburgh, Pennsylvania 15213

INTRODUCTION

The goal of this investigation is to develop a method of producing a nonagglomerating fuel from strongly caking coals. This fuel then could be used in a fluidized-bed gasifier or hydrogenator to make methane. The strongly caking coals, found mainly in Eastern United States where the large market for gas exists, become plastic and cake when heated to gasification temperatures.

An earlier paper described a method of operating with a batch charge of coal in a fluid-bed system.1/ The caking properties of Pittsburgh seam coal and other highly caking coals were destroyed at 400° to 425° C by fluidizing fine coal (18-100 mesh) with steam or nitrogen which contain at least 0.2 percent oxygen. In the batch system a thoroughly nonagglomerating char could be produced in about 5 minutes. The surface of the char was made noncaking in about 1 minute. It was found that the coal must be heated by the fluidizing gas and not through the reactor wall to avoid agglomerating in the pretreating vessel.

The criteria adopted to indicate that the pretreatment was successful in these continuous tests are:

1. Good fluidization must be maintained in the pretreating reactor at the temperature employed.

2. The free-swelling index (FSI) of the resulting char must be less than 2.

3. The char must be noncaking when subjected to a hydrogen treatment at 600° C.

All three conditions must be satisfied before the char is considered noncaking. The free-swelling index, while not a direct measurement of agglomeration properties, does indicate the change in this property. For example, the FSI of Pittsburgh seam coal, a high-volatile A bituminous coal, is about 8, and after successful pretreatment, less than 2.

To optimize the pretreatment, the following information was to be derived from the experiments with continuous feed of coal and discharge of char:

1. The minimum residence time of the coal.
2. The minimum temperature.
3. The minimum oxygen-to-coal feed ratio.
4. The minimum weight loss.
5. The optimum mesh size of coal.
6. The effect of pressure.
7. The quality of the offgas.

APPARATUS AND EXPERIMENTATION

The flowsheet of the continuous unit is shown in figure 1. Nitrogen or steam plus air was used as the fluidizing medium. Coal feed is semicontinuous in that the feeder delivers batches at the rate of about 8 to 20 per minute, producing a feed rate of 120 to 900 grams per hour. This coal is conveyed to the bottom of the reactor by the feed gas. The char is discharged from the lower side arm while the gases, tars, and dusts are discharged from the upper one. The reactor is a stainless steel tube of 1-inch diameter, with the expanded section at the top 2 inches in diameter. The 29-inch section containing the bed of coal is heated electrically. A manometer indicates the pressure drop developed over the coal bed. Because the height of the bed is fixed, the residence time of the coal depends on the rate of coal feed.

DISCUSSION OF RESULTS

More drastic pretreatment was required to destroy the caking properties of coal in the unit with continuous feed and discharge of solids than in comparable batch operations reported previously. Due to the back-mixing of fresh and treated material, some particles remain in the reaction zone a relatively short time and others for a much longer time. Thus the treatment must be severe enough to convert those particle which are in the reactor for a very short time.

Figure 2 shows the results of tests in which the temperature of the bed was varied from 410° to 450° C and the oxygen content of the inert gas used for pretreating was 1.4 and 2.3 percent during pretreatment. Nonagglomerating char was produced in only two of these tests. These employed an oxygen content of 2.3 percent and temperatures of 440° to 450° C, whereas only 0.2 percent oxygen in the fluidizing gas was needed in the batch tests. With a lower oxygen content of 1.4 percent, the chars produced at 440° C caked in the reactor. The higher oxygen content is necessary in the continuous tests because there is a constant feed of raw coal and a greater inventory of coal in the bed tests.

The reason the chars cake during pretreatment at the higher temperatures but not at the lower ones, as demonstrated in figure 2 by the 1.4-percent oxygen parameter, may be explained with the photographs of chars shown in figure 3. At 410° C the particles are discrete, but at 435° C the smaller particles stick to the larger ones. It is believed that more of the viscous, tarry material exudes from the inside of the large particles at the higher temperature. Once the limited amount of oxygen in the gas has been consumed, there is none to oxidize this additional viscous material and the particles stick together. The smaller particles are rendered noncaking more rapidly than the larger particles, so they did not stick to each other, but only to the large particles.

Figure 4 shows the effect on the FSI of varying the residence time from 9 to 34 minutes while holding the oxygen-to-coal ratio constant at about 0.2 cubic foot per pound of coal. Temperature parameters were 420°, 430°, 440°, and 450° C. At this oxygen content, a minimum of about 25 minutes is needed to make noncaking char at 440° C. The time could not be decreased at this oxygen-to-coal ratio by increasing the temperature because at 450° C the char caked in the reactor during treatment. Figure 5 shows the same type plot using an oxygen-to-coal ratio of about 0.3. A residence time of only 7 minutes was adequate to render the coal noncaking at 440° C, and less than 7 minutes at 450° C.

As shown in figure 5, the percentage of volatile matter in the char decreases with increasing pretreatment temperature and residence time. At 14 minutes the volatile matter content of the char is 29 percent at 420° C, and is 23 percent at 450° C. After 34 minutes the volatile matter is 26 percent at 420° C and 21 percent at 450° C.

Since the char from Pittsburgh seam coal is noncaking when its FSI is decreased to 1-1/2, all the data yielding this FSI at 420°, 430°, and 440° C obtained at various residence times and oxygen-to-coal ratios were cross-plotted as shown in figure 6. If a residence time of 5 minutes is desired, the required oxygen-to-coal ratio is about 0.40 when operating at 430° C. Lower temperatures resulted in a much higher oxygen-to-coal ratio or a longer residence time.

Figure 7 shows the effect of temperature on the FSI and volatile matter of char from Pittsburgh seam coal for four different particle sizes at a constant residence time of 14 minutes. The oxygen-to-coal feed ratio was constant at 0.3 cubic foot oxygen per pound of coal. For the 18-48 mesh size the FSI decreased rapidly from 6-1/2 at 410° to 1 at 440° C. Lower free-swelling values were obtained with decreasing sizes of particles treated at similar temperatures; for example, for the 150-200 mesh the indices were 1 at 410° C and noncaking (NC) at 450° C. These results show how much more readily the finer coal sizes can be pretreated.

Loss of volatile matter is more rapid for the small particles than the coarser sizes. Also, as anticipated, the loss of volatiles increases with increasing temperatures. The loss of volatile matter of 18-48 and 48-100 mesh sizes for satisfactory decaking of coal is about 10 percent. This is roughly equivalent to the weight loss and to the loss in heating value of the coal.

Since the finer size particles require less drastic treatment, an attempt was made to show that the oxygen needed for pretreatment varied with the particle diameter. As shown in figure 8, the oxygen-to-coal ratio varied from 0.42 for the 18-48 mesh to 0.06 for the 150-200 mesh. This theory was partially successful, but the finer mesh sizes caked in the reactor when the bed temperature was raised above 430° C. The mesh sizes finer than 100 mesh were more difficult to fluidize.

Figure 9 shows the effect of temperature on the quality of offgas made during pretreatment with steam plus air. The oxygen-to-coal feed ratio was 0.4 cubic foot per pound. The quantity of methane and higher hydrocarbons increased with increasing temperatures. The yields of carbon oxides increased slightly, but the hydrogen yield remained constant at about 0.2 cc/gram. Similar results were obtained in the batch tests except that the yield of hydrogen (about 66 cc/gram in the batch tests) is lower in the continuous operation, probably because the oxygen needed to treat the coal reacted with the hydrogen.

Table 1 shows analysis of coal and chars used in the above tests. As in the batch tests, the oxygen content is greater in the raw coal than in the chars.

TABLE 1.- Analysis of coal and chars from the tests to study the effect of temperature on the offgas made during steam-air treatment of Pittsburgh seam coal of 48-100 mesh size

Analysis (as received)	Coal	Chars 410° C	420° C	430° C	440° C	450° C
Proximate, percent						
Moisture	1.5	0.1	0.1	0.3	0.0	0.3
Volatile matter	36.0	27.2	26.9	25.9	23.4	22.7
Fixed carbon	54.4	64.4	64.6	66.9	68.6	67.9
Ash	8.1	8.3	8.4	6.9	8.0	9.1
Ultimate, percent						
Hydrogen	5.2	4.5	4.4	4.4	4.2	4.1
Carbon	75.2	77.3	76.9	78.0	77.0	76.1
Nitrogen	1.5	1.6	1.3	1.6	1.6	1.6
Oxygen	7.9	6.4	7.1	7.4	7.4	7.2
Sulfur	2.1	1.9	1.9	1.7	1.8	1.9
Ash	8.1	8.3	8.4	6.9	8.0	9.1
Heating value, Btu/lb	13,410	--	13,240	13,470	--	13,130

The effect of pressure was studied at 1, 5, 10, and 20 atmospheres using steam plus air as the fluidizing gas. A mesh size of 150-200 was used so that a lower fluidization gas velocity could be employed. Generally there was no great change due to pressure. Using a fluidizing gas mixture of steam plus air with an oxygen-to-coal ratio of 0.3 cubic foot per pound at 430° C, as the pressure was increased the yield of carbon oxide gases decreased, but the hydrocarbon yield increased. The amount of coal throughput increased as the pressure was increased, but at a rate less than linearly. Again, as in the batch tests, the particles seemed to explode when the pressure was released, probably because trapped gas in the particles escaped on depressurization.

CONCLUSION

The tests have demonstrated the operability of a small-scale continuous unit designed to pretreat highly caking coal to remove its caking quality. The principal difference between results of continuous and batch operation is the need for a higher oxygen content of the feed gas in continuous flow. The minimum residence time required for the coal to be in the reactor is about 5 minutes at 430° C. About 0.4 cubic foot of oxygen is needed in the treating gas for each pound of coal to destroy the caking quality of Pittsburgh seam coal. Although the optimum mesh size is 18-100, finer mesh sizes permit less drastic treatment. However, finer mesh sizes are more difficult to fluidize. Minimum weight loss was 10 percent, which is equivalent to 10 percent loss of heating value of the original coal. The effect of pressure is negligible compared to atmospheric pressure. However, higher throughputs can be achieved at higher pressures.

Steam plus oxygen is the desired fluidizing medium if both the char and the offgas are to be utilized in a gasifier. This results in an advantage over other methods of treatment because the volatile matter evolved during the pretreatment is not removed from the system.

Reference Cited

1. Forney, A. J., R. F. Kenny, S. J Gasior, and J. H. Field. The Destruction of the Caking Properties of Coal by Pretreatment in a Fluidized Bed. Ind. and Eng. Chem. Product Research and Development, v. 3, No. 1, March 1964, pp. 48-53.

3-17-64 L-8524

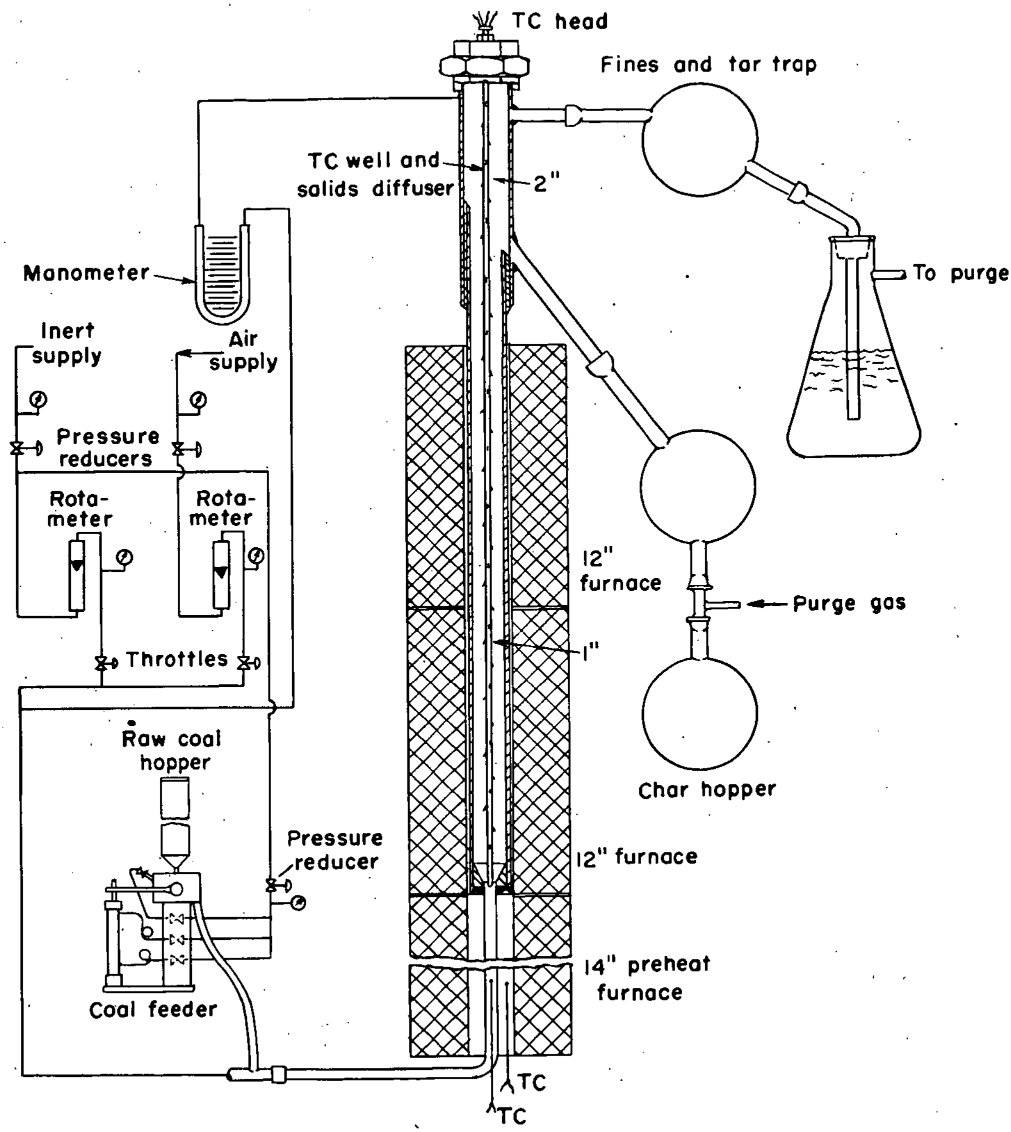

Figure 1. Continuous fluidized-bed coal pretreater.

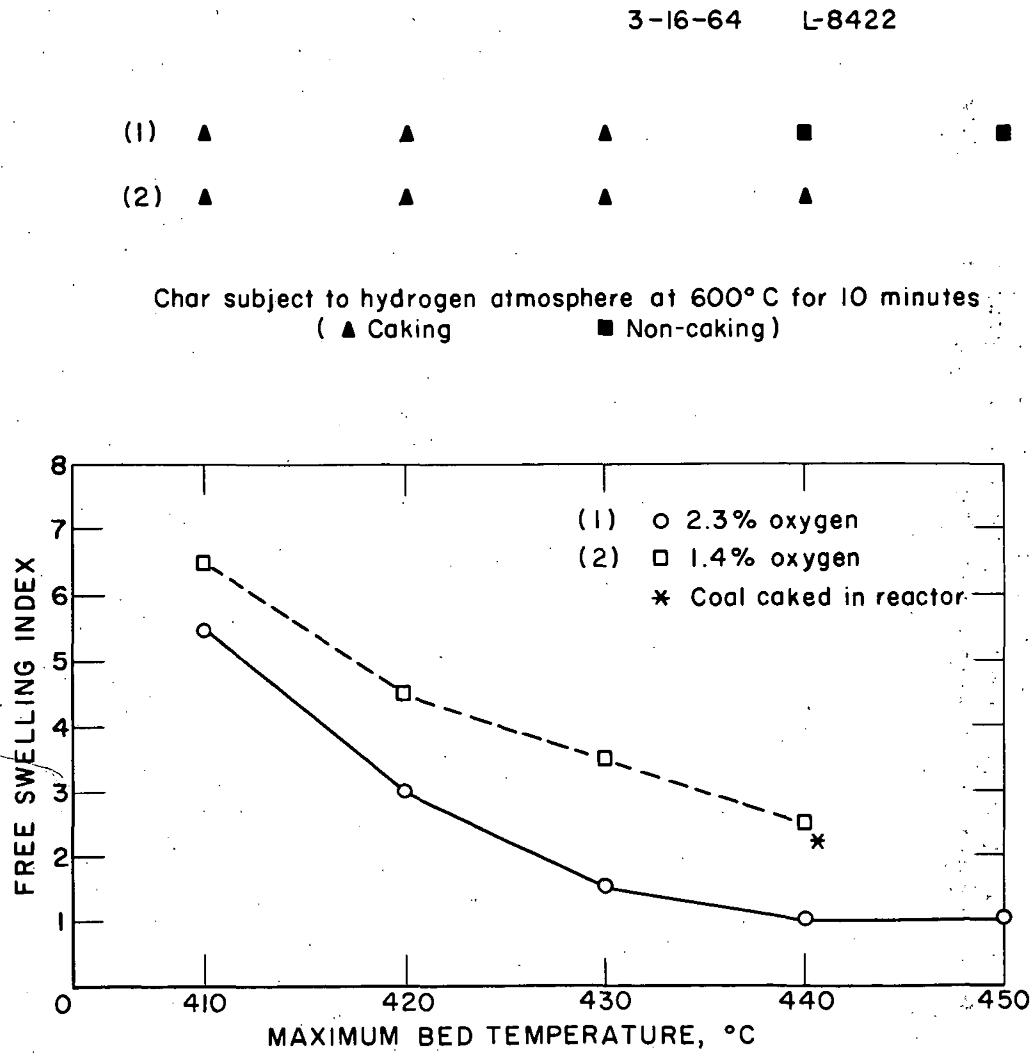

Figure 2. The effect of oxygen content of the fluidizing gas and of temperature on the caking properties of Pittsburgh seam coal (18-100 mesh). The fluidizing gas is inert gas with oxygen content as noted. The average residence time of the char in the reactor is 19 minutes.

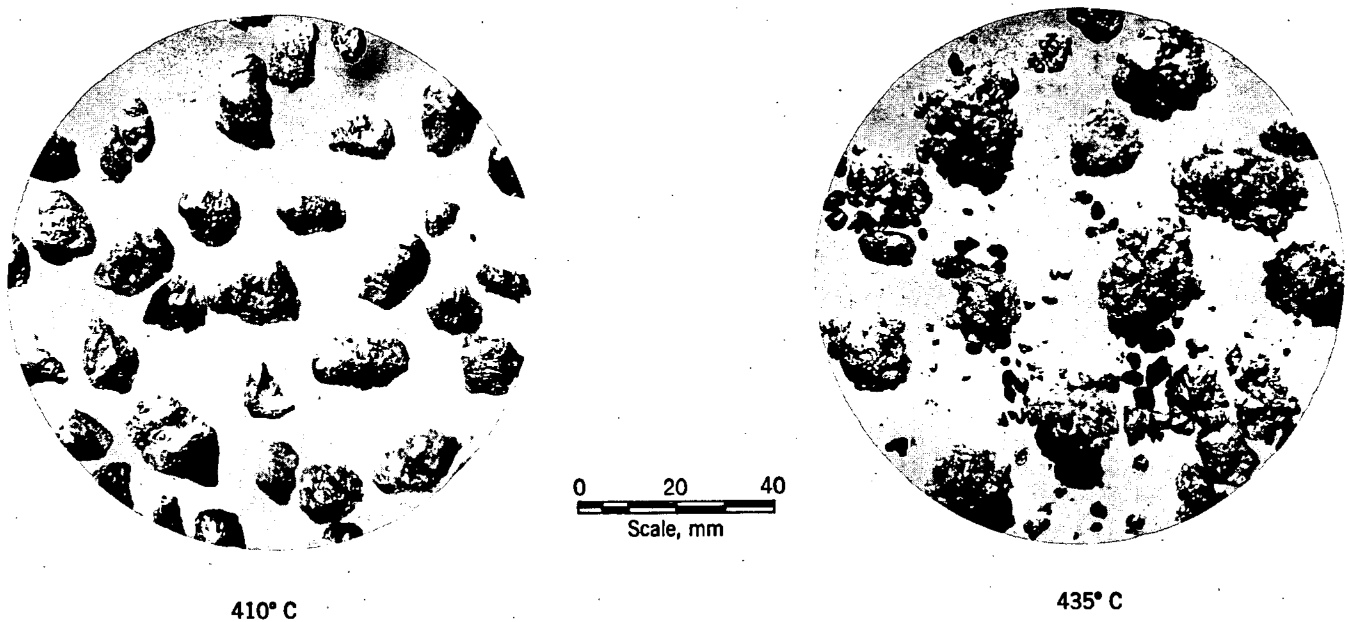

Figure 3, The effect of increased temperature on the caking property of char made from Pittsburgh seam coal (18-100 mesh), 1.4 percent oxygen in the feed gas.

H-91086-P

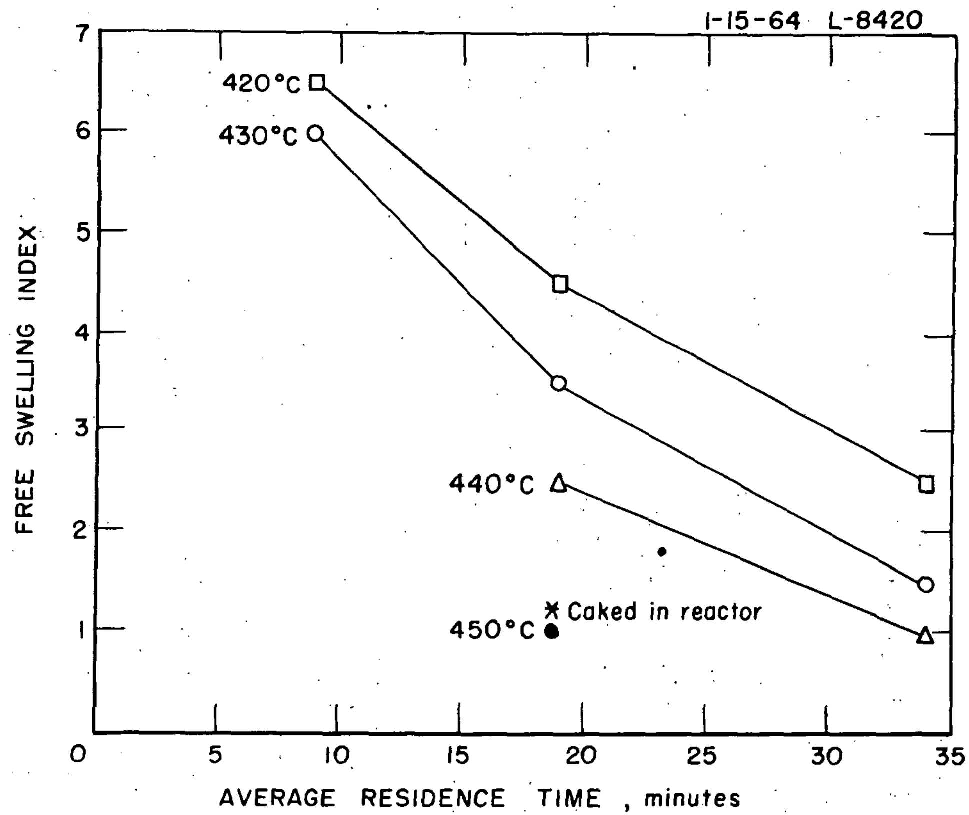

Figure 4. The effect of residence time and of temperature of pretreatment on the free-swelling index of char from Pittsburgh seam coal (18-100 mesh) 0.18-0.20 ft.^{3} oxygen/lb coal feed.

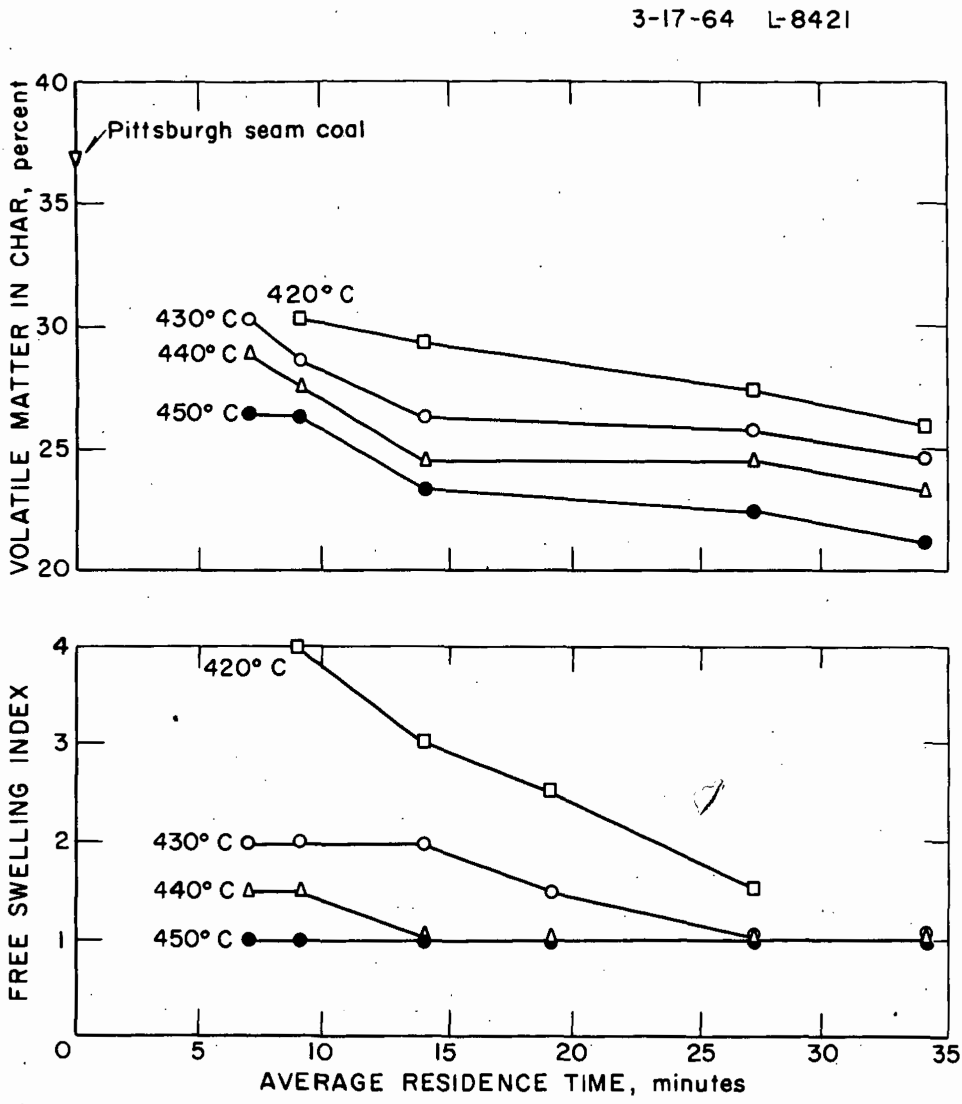

Figure 5. The effect of residence time and of temperature of pretreatment on the free-swelling index and the volatile matter of the char from Pittsburgh seam coal (18-100 mesh; 0.28-0.33 cu ft oxygen/lb coal feed).

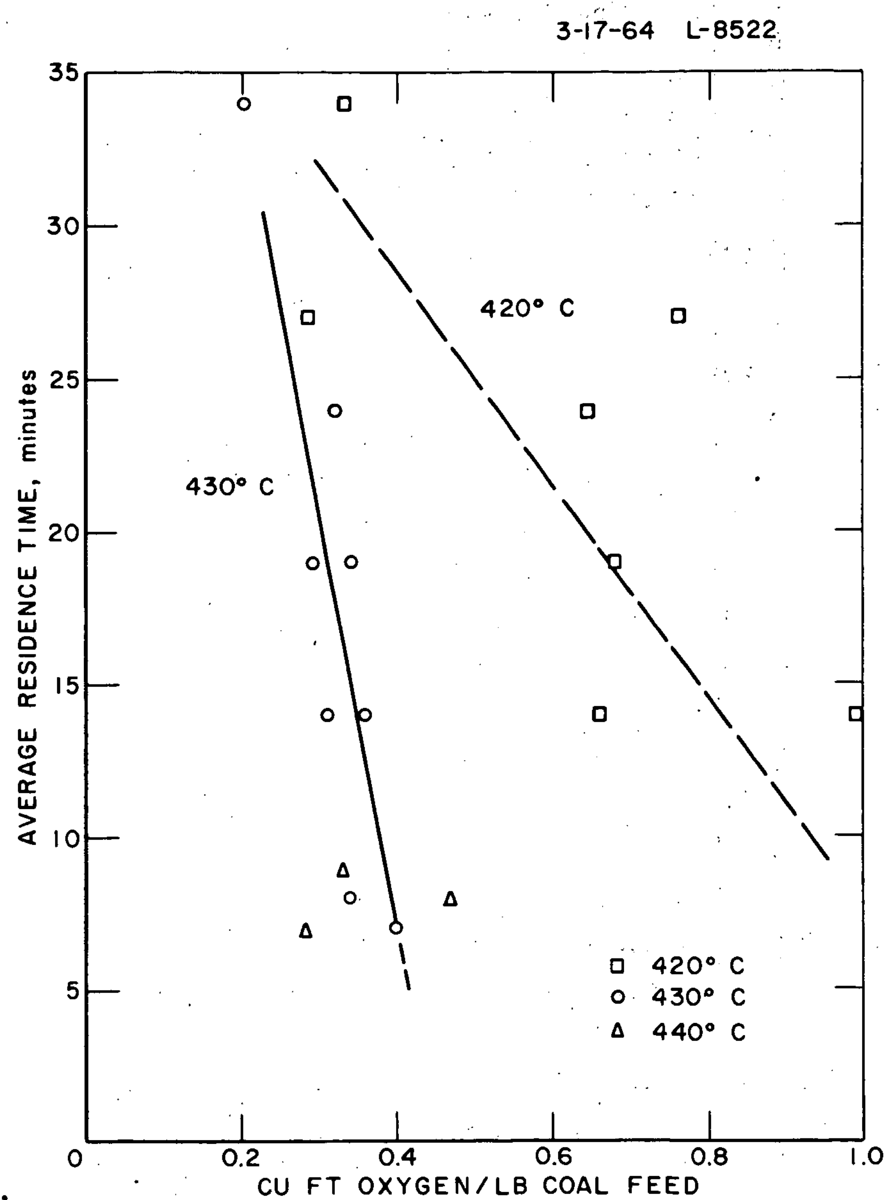

Figure 6. The effect of temperature and oxygen-coal ratio on the residence time of Pittsburgh seam coal of 18-100 mesh (FSI = 1-1/2).

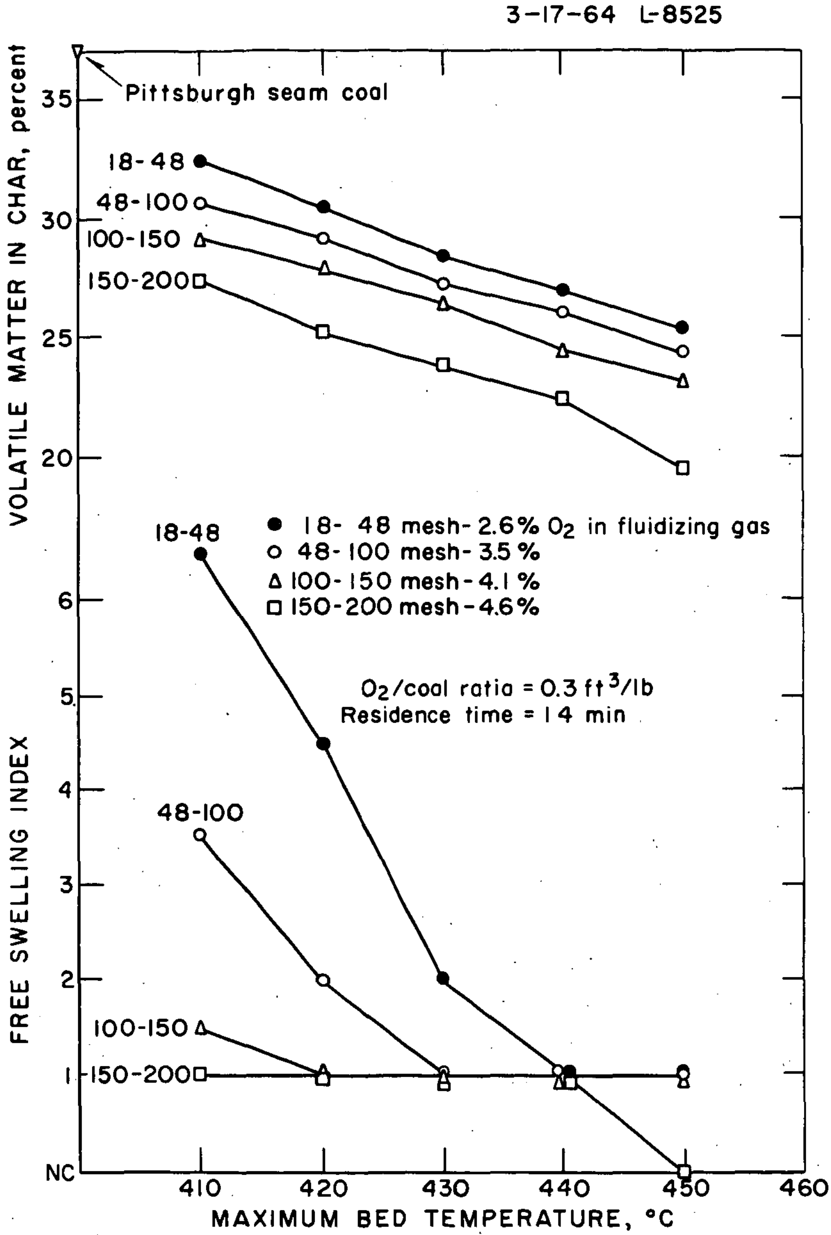

Figure 7. The effect of mesh-size and temperature on the free-swelling index and volatile matter content of the char made from Pittsburgh seam coal.

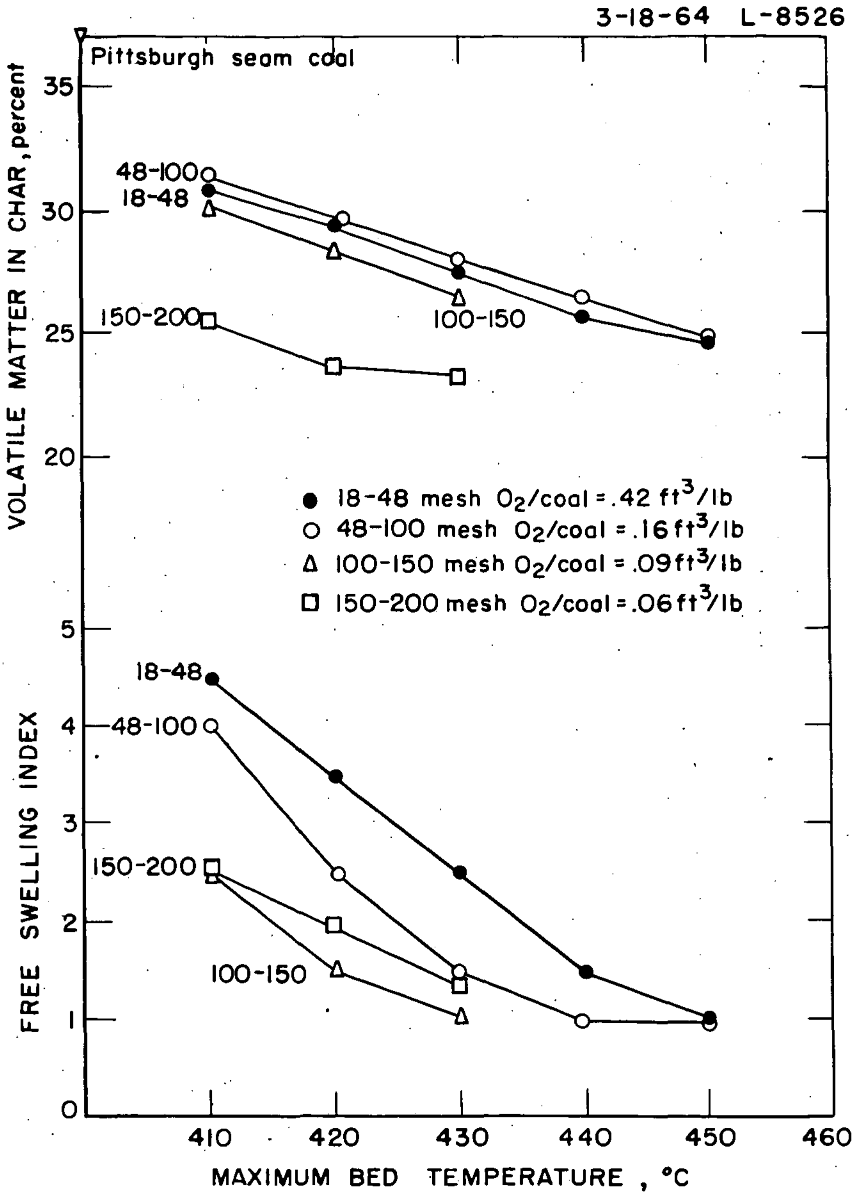

Figure 8. The effect of mesh size and temperature on the free-swelling index of char made from Pittsburgh seam coal. (The oxygen/coal feed ratio varied as noted; residence time is 14 minutes.)

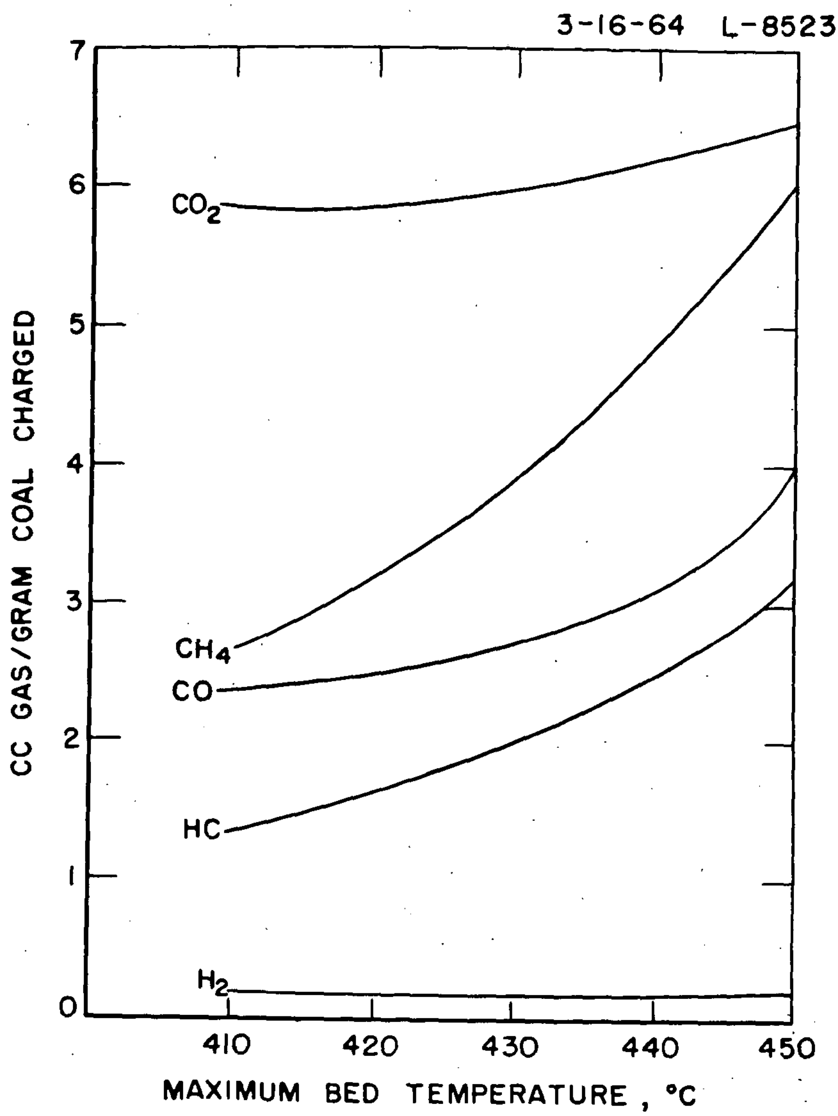

Figure 9. The effect of temperature on off gas made during steam-air treatment of Pittsburgh seam coal of 48-100 mesh (O_2-coal ratio = 0.4 cu ft/lb; residence time is 14 minutes).